

CONTRIBUTION OF ACTIVATION PRODUCTS TO FUSION ACCIDENT RISK: PART II. EFFECTS OF ALTERNATIVE MATERIALS AND DESIGNS

JOHN P. HOLDREN *University of California, Berkeley*
Energy and Resources Group, Berkeley, California 94720

and

University of California, Lawrence Livermore National Laboratory
P.O. Box 808, Livermore, California 94550

STEVE FETTER *University of California, Berkeley*
Energy and Resources Group, Berkeley, California 94720

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Comparison of accident-hazard potentials associated with neutron-activation products in fusion reactors of various designs and structural materials suffers from a number of shortcomings in the readily available hazard-index data. Neither inventories of curies nor biological hazard potentials (BHPs) are satisfactory indices of hazard even if consistently computed, and between-study inconsistencies in neutronics packages and BHP calculations further obscure the meaning of comparisons based on these measures. We present here the results of internally consistent calculations of radioactive inventories, BHPs, and off-site dose potentials associated with the first walls of nine reactor-design/first-wall-material combinations. A recent mirror-reactor design reduces off-site dose potentials by a factor of 2 compared to a much-studied early tokamak, for a given first-wall material. Holding design fixed, HT-9 ferritic steel offers a factor of 2 reduction in dose potential compared to Type 316 stainless steel. By the dose-potential measure, molybdenum is the worst of the materials investigated and silicon carbide is by far the best. Hazards in realizable accidents depend not only on the hypothetical dose potentials, as calculated here, but also on the actual release fractions of first-wall (or other activated) material. Review of the theoretical and experimental evidence bearing on release fractions suggests that, for most candidate materials, high release fractions from designs containing liquid lithium cannot yet be convincingly ruled out.

I. INTRODUCTION

The hazards associated with activation-product inventories in fusion reactors are mainly of three kinds: irradiation of workers during routine operation, maintenance, and decommissioning; irradiation of workers and the public by activation products released in severe reactor accidents; and escape of radionuclides from waste repositories, with the potential to irradiate members of the public in this generation or future ones. The magnitudes of these hazards are susceptible to reduction to some extent by means of two general approaches: reducing the inventories of troublesome activation products (either by the choice of fusion fuels and structural materials, or by reactor designs that minimize the interaction of neutrons with the structural materials); and controlling the potential pathways to release/exposure through engineered safeguards and suitable operating procedures.

Measures taken to reduce hazards may impose performance penalties and additional costs, however, and measures intended to reduce one set of hazards may aggravate others. Such hazard-abatement approaches as the use of low activation structural materials, if deemed to be essential, might even delay the availability of commercial fusion energy by posing additional development and engineering problems. To determine whether a given hazard-reduction measure is worthwhile, it is necessary to weigh the *size* of the reduction against the costs incurred (including any increases in other hazards and delays in the commercialization of fusion). Clearly, neither substantial costs nor significant delays should be incurred

to obtain only a small reduction in a large hazard, or to reduce by a large fraction a hazard that is minor already. On the other hand, measures that can greatly reduce hazards large enough to imperil the commercial attractiveness or public acceptability of fusion are likely to be worth their costs in money and time.¹

As part of the broader analysis necessary to illuminate this issue, we have been examining quantitative measures of the hazard to public health from postulated severe accidents in deuterium-tritium-fueled fusion reactors with various structural materials and designs. In a predecessor to the present article,² it was shown that, for the UWMAK-I fusion-reactor design with stainless steel, molybdenum, or niobium structure, release of those first-wall activation products that form oxides volatile at temperatures attainable in lithium fires could produce off-site radiation doses comparable to those predicted for severe fission-reactor accidents. Sawdye and Kazimi³ reached similar conclusions about the hypothetical dose potential of the activation products in stainless steel and molybdenum first walls, but they argued that the large release fractions necessary actually to deliver such doses are implausible, at least in the stainless steel case.^{3,4}

In the present paper, we extend our previous work to explore indices of accident hazard for additional reactor designs and structural materials. In so doing, we uncover a number of obstacles to meaningful safety comparisons based on the information in existing fusion-reactor design studies, we describe some improvements in our own models for carrying out such analyses, and we review in some detail the available evidence bearing on the crucial question of release fractions.

II. INDICES OF POTENTIAL HAZARD

II.A. Types of Indices

The literature of fusion-reactor hazards makes use of a number of different indices of the potential hazard associated with accidental release of neutron-activation products. The three most common units of measurement for such indices are (a) curies of radioactivity ($1 \text{ Ci} = 3.7 \times 10^{10} \text{ dis/s}$); (b) cubic metres of biological hazard potential (BHP)—more informatively termed “dilution volume,” based on its calculation as the sum of the volumes of air needed to dilute each radionuclide present to its maximum permissible concentration (MPC), as specified for public exposure in the Code of Federal Regulations⁵; and (c) rems of radiation dose hypothetically deliverable to an individual member of the public

(or person-rems of population dose, obtained by summing individual doses over the entire population hypothesized to have been exposed).

Different indices are obtained, moreover, depending on whether these quantities are based on the activation products contained in the coolant alone, the first wall alone, the whole blanket, or the whole reactor; and on whether the full inventories are taken into account or only those fractions postulated to be releasable under physically realizable conditions. And, finally, the magnitudes obtained depend on the power level and duration of operation assumed, and on the time period after reactor shutdown for which the assessment is made.

II.B. Indices in Previous Studies: Curies and BHPs

The major reactor-design studies, for example, UWMAKs I, II, and III (Refs. 5, 6, and 7), NUMAK (Ref. 8), WITAMIR (Ref. 9), and Starfire (Ref. 10), typically have provided data for whole-blanket and coolant inventories of curies, with partial breakdowns by blanket region and by isotope, based on the use of large computer codes and nuclear-data libraries to perform the necessary neutron-transport, activation, and decay calculations. Most of these studies also have provided some corresponding figures for BHPs, although not for hypothetical radiation doses due to accidental releases. (One early study that did calculate off-site radiation doses from a postulated release was the environmental assessment for the “Brookhaven Minimum Activity Blanket.”¹¹)

While the main reports of most of the major reactor-design studies present activation-product inventories only for a single candidate structural material, data have long been available showing how radioactive inventories and BHPs vary with the choice of material for at least a few designs (see, e.g., Refs. 6 and 12 through 15). One early comparative study varied materials and reactor designs simultaneously, presenting inventories and BHPs for five combinations of design and materials choice: four tokamak designs with structures of stainless steel, nickel alloy, niobium, and aluminum/aluminum-oxide respectively; and a stainless steel mirror reactor.¹⁶ These early comparisons of numbers of curies and BHPs did much to shape perceptions about the relative activation hazards of different materials choices.

II.C. Indices in Previous Studies: Off-Site Doses

The U.S. Nuclear Regulatory Commission's (NRC's) Reactor Safety Study of 1975, treating probabilities and consequences of large releases of radioactivity from light-water-moderated fission power reactors, improved and called widespread attention to the calculational tools for estimating the doses to people that would result from given

^aThus, the BHP is given by $\sum N_i / \text{MPC}_i$, where N_i is the number of curies of the i 'th isotope and MPC_i is the maximum permissible concentration for that isotope, in Ci/m^3 .

releases of radioactive material.¹⁷ These tools included modified Gaussian-plume models for calculating the atmospheric dispersion and ground deposition of airborne materials, external-dosimetry models for deriving gamma-ray doses from air and ground concentrations of radionuclides, and internal-dosimetry models for calculating the doses from inhaled and ingested radionuclides. An early application of such methods to fusion activation products was the work of Clark,¹⁸ who calculated off-site doses per released curie of nine structural isotopes in addition to tritium, ^{14}C , and ^{41}Ar .

Subsequently, Kazimi and Sawdye^{3,19} used similar tools to calculate off-site population doses for postulated releases of activated first-wall material from two conceptual tokamak reactors—UWMAK-I [Type 316 stainless steel (316 SS)] and UWMAK-III (TZM molybdenum alloy). Their published reports on this work, however, do not present the doses themselves but instead show a more intricately derived and far less transparent result: the limiting frequency distribution of releases of various fractions of the fusion first walls, such that the expected harm (early fatalities and illnesses and latent cancer fatalities per year of reactor operation) does not exceed that predicted for light water reactors by the Reactor Safety Study.

In the predecessor to the present paper, Holdren² used similar consequence models to estimate the potential radiation dose at the plant boundary (assumed 200 m from the release point) for postulated severe releases from UWMAK-I reactors using three different structural materials: 316 SS, TZM molybdenum alloy, and NbZr niobium alloy. That paper shows the contributions of individual isotopes to the boundary dose and compares this set of indices to some simpler ones: numbers of curies and BHPs weighted by the same postulated release fractions as were used in the dose calculations.

Probably the most ambitious exploration of hazard indices for fusion accidents available to date is the 1982 doctoral dissertation of Piet,²⁰ which presents a number of indices related to the chances of a large release of activation products as well as indices related to potential consequences. Piet considered 11 coolant/breeder/structure variants of the Starfire tokamak design, involving four different structural materials (316 SS, TZM molybdenum alloy, V-15 Cr-5 Ti vanadium alloy, and HT-9 ferritic steel). He presents radioactivity inventories for only the first three materials, however, and, while he manifestly has calculated both early and chronic off-site doses from postulated releases of these materials, he never presents the doses themselves: The reader sees only a set of derivative indices—the doses expressed as multiples of those from the least hazardous material (the vanadium alloy) and the fractions of the inventories that would have to be

released to produce early fatalities and injuries at an “average” site under “average” weather conditions.

III. OBSTACLES TO MEANINGFUL COMPARISONS

III.A. Radioactivity Inventories

The most widely available index of potential accident hazard associated with activation products—the number of curies present after a specified period of operation at a given power level—is also the least informative. Different isotopes differ so drastically in such hazard-determining properties as half-life, decay mode and energy, and radioactive daughter products that aggregated curie totals for different systems convey next to no information about relative hazard. If one happens to know, for a given structural material, which few among the many isotopes produced by neutron activation dominate the overall hazard, then one can compare the curie inventories of these isotopes, in different reactor designs using the given material, to get an indication of the relative activation hazards associated with these designs. But knowing which isotopes are the critical ones requires reference to the more sophisticated hazard indices.

Two further obstacles impede the use of calculated inventories of curies both for direct hazard comparisons and as the starting point for calculating more informative indices.

First, few of the reports of reactor studies in which activation-product inventories were calculated present the results with the degree of disaggregation appropriate for hazard comparisons. Some of these reports disaggregate the numbers of curies to the level of elements but not to the level of individual isotopes (telling one, for example, how many curies of niobium isotopes are present but not how these are distributed among ^{92}Nb , ^{94}Nb , ^{95}Nb , and so on); some do not give even this much information; and almost none specify what fractions of the inventories, disaggregated by isotopes, are found in the first wall versus in the coolant, the rest of the blanket, and the shield. (Disaggregation of this last sort is essential, of course, if plausible release fractions are to be accounted for in the construction of more informative indices.) Alas, even the most recent studies are no exception to this problem: The Starfire report¹⁰ disaggregates total curies by blanket regions but does not provide usable isotopic breakdowns; Piet's dissertation²⁰ presents—and derives other hazard indices from—inventories “equivalent to 5 millimeters of first wall material,” although the first-wall thicknesses employed in the underlying neutronic calculations differ among materials and are all <5 mm.^b

^bPiet's inventories are therefore neither fish nor fowl: They represent more curies than are in the first wall (by a margin that varies from one material choice to another) and fewer than are in the blanket as a whole (again by a variable margin).

Second, comparison of inventories of curies as calculated by different reactor-design groups, or even as calculated for successive designs by the same group, can be misleading because of noncomparability in the neutronics codes and cross-section libraries used for these calculations. The various neutronics codes contain assumptions and approximations, the full implications of which may be known only to the groups that wrote them; and a single reactor design, structural material, and library of activation cross sections may yield significantly different results for activation-product inventories when analyzed with different codes. Given that there is not one accepted library of activation cross sections but several—containing some significant discrepancies and all in a more or less constant state of revision—the problem of inventory comparisons based on published studies is even greater. (Sophisticated techniques exist for analyzing the uncertainties in the results of a neutronic calculation that result from uncertainties in the input nuclear data,^{21,22} but we have seen no analyses applying these techniques to activation-product-inventory calculations.) As a result, it can be extremely difficult or impossible to determine whether the observed differences in inventories are due to differences in reactor design and materials choices, on the one hand, or to differences in the calculational schemes and cross-section libraries, on the other.

III.B. Biological Hazard Potentials

The use of BHPs for hazard comparisons cannot avoid most of the shortcomings we have just attributed to inventories of curies, since isotope-specific knowledge of the inventories is an essential input to the calculation of BHPs. Where that knowledge is available and reliable, however, taking the next step and calculating and comparing BHPs does, in principle, offer more insight than working with curies. The BHP's advantage as a hazard index is that it incorporates, through its dependence on the MPCs of the isotopes present, a substantial amount of hazard-related information about half-lives, decay energies, and dosimetry.

The usefulness of hazard comparisons based on the BHP figures available in the fusion-reactor literature is less than might be expected, however, even discounting the problems with the underlying curie figures. A key additional problem has been the treatment of the many activation products for which the Code of Federal Regulations lists no MPCs. In most cases, the contributions of such isotopes to the BHP either were ignored or were approximated (badly) by use of the "default" MPC value suggested in the regulations for unlisted isotopes. In other cases, MPCs for the unlisted isotopes have been estimated by analogy to listed isotopes having similar decay modes

and half-lives; and a few authors have used prescriptions similar to that of the International Council on Radiation Protection and Measurements (ICRP), which is the source of the officially listed values, to derive MPCs for some of the most conspicuous unlisted fusion-activation isotopes.

Recently this problem was addressed more systematically by Fetter: His report²³ presents a computer code that calculates MPCs using the ICRP's approach, given the decay scheme of the isotope and basic information about the physiological behavior of the chemical element to which it belongs. The code was tested by using it to reproduce the MPCs of 78 officially listed isotopes; the report presents these results together with calculations of MPCs for 53 unlisted isotopes of particular fusion interest. This code provides the capability to carry out BHP comparisons that are as good as the curie-inventory data they use as input.

III.C. Potential Doses versus BHPs

BHPs, consistently calculated, provide a better index of potential hazard than do raw data on curies, but they are not as illuminating as information on the radiation doses potentially deliverable to people off site. One apparent reason for preferring a dose calculation is that the BHPs do not account for how much of the radioactive inventory actually could be released. This seeming disadvantage of the BHP index actually is illusory, however. If release fractions can be estimated, then the BHP of each isotope can be multiplied by the appropriate fraction to give a release-weighted BHP. (This procedure was used in Part I of the present study.²) If the possible release fractions are completely unknown, on the other hand, then the dose index is handicapped in the same way and to the same degree as is the unweighted BHP index. For the only meaningful dose calculation one can do in that case is of the "dose potential" associated with the full inventories of various parts of the reactor—the first wall, the coolant, the whole blanket—analogue to the unweighted BHPs.

The reason for preferring radiation dose to BHP as an index of hazard, then, has to do not with release fractions but with several other factors. One is that the units of dose—say, rem—are more transparent as an index of the magnitude of the potential hazard than are the units of BHP—say, cubic kilometres. Another factor is that the MPCs (which, together with the inventories, determine the BHPs) are derived on the basis of continuous exposure via inhalation or ingestion of fluids (including the water content of food); external irradiation is ignored except in the cases of the noble gases, for which the MPCs in air are derived for continuous exposure in a uniformly contaminated atmosphere. In an actual accident, any doses capable of producing early

fatalities and illnesses (as opposed to delayed cancers) would result from transient exposure, via inhalation and external irradiation, to a highly nonuniform passing cloud of radioactivity and the ground contamination it left behind. The external and internal dosimetry will be different enough in the two cases that, on this ground alone, the MPC-based BHPs and the calculated doses for an accident situation are likely to differ substantially in the picture they convey of the relative importance of different isotopes and the comparative hazards of different reactors. Other factors that contribute to the difference between the relative-hazard measures provided by BHPs and doses are (a) the inflation in aggregated BHPs resulting from the "most sensitive organ" dependence of the MPCs, (b) some outdated "correction" factors in some of the ICRP's models for MPC calculation, and (c) an artificial compression of between-isotope BHP differences that results from the convention of using the lower of the "soluble" and "insoluble" MPCs.^c

The foregoing points make a strong case for some form of off-site dose potential as the most meaningful index for comparing the hazards of different fusion reactor designs and materials. Another point in its favor is that calculating off-site doses is not much more difficult or consumptive of computer time than is calculating BHPs—certainly not in cases where the latter involves derivation of MPCs of isotopes without listed values. (The extra work in the dose calculation is in the dispersion models, finite-cloud dosimetry corrections, and such; these aspects can be tedious to code but, once that is done, they run quickly. The dominant consumer of computer time in the calculation of *any* of the hazard indices we are considering here is the use of large neutronics codes and cross-section libraries to obtain the inventories of isotopes.)

III.D. Difficulties with Doses

There are, nonetheless, some penalties associated with choosing to work with doses as one's index of hazard. The addition of another set of models, beyond those needed for a BHP calculation, adds opportunities for different analysts to make different approximations and assumptions. For example, Clark¹⁸ and Holdren² considered the initial external and inhaled doses from the passing cloud, but not the dose from radioactive material deposited on the ground; Kazimi and Sawdye¹⁹ included the initial

ground dose as well, requiring further assumptions about how long people remain in the contaminated area; and Piet²⁰ included all this plus chronic doses from long-term contamination of ground, water, and food, requiring still more choices of models and parameters. Different ways to treat building-wake effects, thermal plume rise, and radioactive self-heating of the plume abound in the literature. And different analysts may choose to calculate doses under different weather conditions (atmospheric stability, inversion height, wind speed, and so on), at different distances from the accident, with different assumptions about holdup time between reactor shutdown and the release of radioactivity to the environment.

All these factors can be held constant, of course, for any one comparative investigation in which all the calculations are conducted from scratch. For purposes of comparing materials and designs, *consistency* of assumptions and approximations in the dose modeling matters more, in general, than the particular choices of assumptions and parameters: the comparative size of the doses from different reactor designs and materials choices can be meaningful and informative even where modeling uncertainties make the absolute values highly suspect. (The choice of assumptions and parameters *may* influence the relative standing of different materials, however, in cases where much of the hazard of one of the materials is associated with only one or two isotopes.) The main problem is that meaningful dose comparisons drawn from *different* studies in the literature will not be possible unless and until investigators agree on a common set of assumptions and parameters for such estimates (even assuming that inconsistencies arising from neutronics codes and cross-section libraries are resolved). Otherwise, there will remain no way to know which differences are due to the models and which to the designs and materials being compared.

IV. SOME NEW COMPARISONS

In this section, we describe some calculations we have carried out to illustrate and to circumvent at least partly the above-described obstacles to meaningful comparisons of hazard indices for fusion reactors. This work has consisted of (a) assembling and testing a set of suitably comprehensive and consistent computational models for calculating radioactive inventories, BHPs, and off-site dose potentials, and (b) applying these models to two conceptual fusion-reactor designs and seven materials choices in a way that permits comparisons among indices, designs, and materials, and between the new and previous calculations.

^cThe compression results because the minimum MPC for many isotopes is determined by the lung dose delivered by the insoluble forms, which dose depends only on decay energy and half-life, but not on the chemical characteristics so important in determining the doses to the rest of the body.

IV.A. Models and Data Libraries

IV.A.1. Neutron Transport and Activation

Neutron-transport calculations were carried out using the TART code,²⁴ which was developed at the Lawrence Livermore National Laboratory (LLNL). Cross sections from the ACTL library²⁵ (also developed and maintained at LLNL) were integrated over the TART fluxes with the ORLIB code²⁶; the results were then supplied to the FORIG code²⁷ (an LLNL adaptation of Oak Ridge National Laboratory code,²⁸ ORIGEN2), which carried out the activation and decay calculations leading to the inventories of radionuclides in fusion-reactor structures. These steps were all performed on the Cray-I at the National Magnetic Fusion Energy Computing Center at LLNL. The structure of this calculation is shown in Fig. 1.

Both of the blanket designs studied were modeled assuming cylindrical geometry. When alternative first-wall materials were considered, they were substituted for the original first-wall material on a 1:1 volume basis throughout the blanket. The Monte Carlo transport calculations were carried out using 10 runs of 1000 neutrons each; the resulting standard deviations in the path length and energy transported into the first-wall zone were on the order of 1.5% in all cases.

That the results of such calculations can indeed depend on which of the available neutron-transport codes, cross-section libraries, and activation codes are used is illustrated by Table I. It compares (a) the inventories we calculated for the UWMAK-I (316 SS) first wall using TART/ACTL/FORIG with the results obtained for the same case by the University of Wisconsin Group in 1974 using the ANISN neutron-transport code, the ENDF/B-III cross-section library⁵

and the DKR activation code, and (b) TART/ACTL/FORIG inventories for the Mirror Advanced Reactor Study (MARS) baseline first wall (HT-9 ferritic steel) with ANISN/ENDF/B-IV/DKR results for the same case.²⁹ (Only isotopes with inventories above 10^5 Ci are shown in the table. Isotopes listed as omitted by one or the other of the neutronics packages were not calculated at all by that package.) The differences in the computed inventories are a factor of 2 or more for 5 of the 18 isotopes calculated for UWMAK-I by both packages and greater than a factor of 3 for 9 of the 22 isotopes calculated for MARS by both packages.

We suspect that the main differences arise from the libraries rather than from the neutron-transport or activation codes, and we believe that the ACTL library, which was developed for fusion applications, has a more complete and more accurate set of cross sections for these purposes than does the more fission-oriented ENDF library. The main point, however, is that more systematic comparison and reconciliation of the different neutronics packages needs to be carried out. Otherwise it will remain impossible to draw meaningful conclusions about comparative hazard measures when the configurations involved have not all been analyzed with the same package.

IV.A.2. BHP Calculations

Biological hazard potentials were calculated using MPCs for continuous public exposure to contaminated air. The usual procedure of using the lower of the two MPCs (for soluble and insoluble forms of each isotope) was followed. In the case of isotopes for which no MPCs are listed in the Code of Federal Regulations,³⁰ MPCs were calculated using the computer code developed by Fetter.²³ This code, which is written in FORTRAN 77 for the Lawrence Berkeley Laboratory's VAX-11 computer, calculates each MPC based on computed doses to the whole body and to 30 individual organs. It uses absorption and retention factors for each element from the original ICRP data base³¹ and decay schemes and energies from the *Table of Isotopes*, 7th edition.³²

Table II illustrates how the use of meaningful MPC values—as opposed to “default” values—for isotopes not officially listed can influence both the absolute magnitude of BHP figures and the relative sizes of BHPs for different materials. For four different materials choices for a MARS first wall, the table shows some unlisted isotopes for which use of the default MPC values produces a substantial difference in the aggregate BHP associated with the first wall. The BHP contributions of these isotopes are shown for three circumstances: MPCs calculated²³ using the ICRP's model; MPCs equated to the “default” value for unlisted isotopes; and MPCs taken from an earlier comparative study¹⁴ in which some

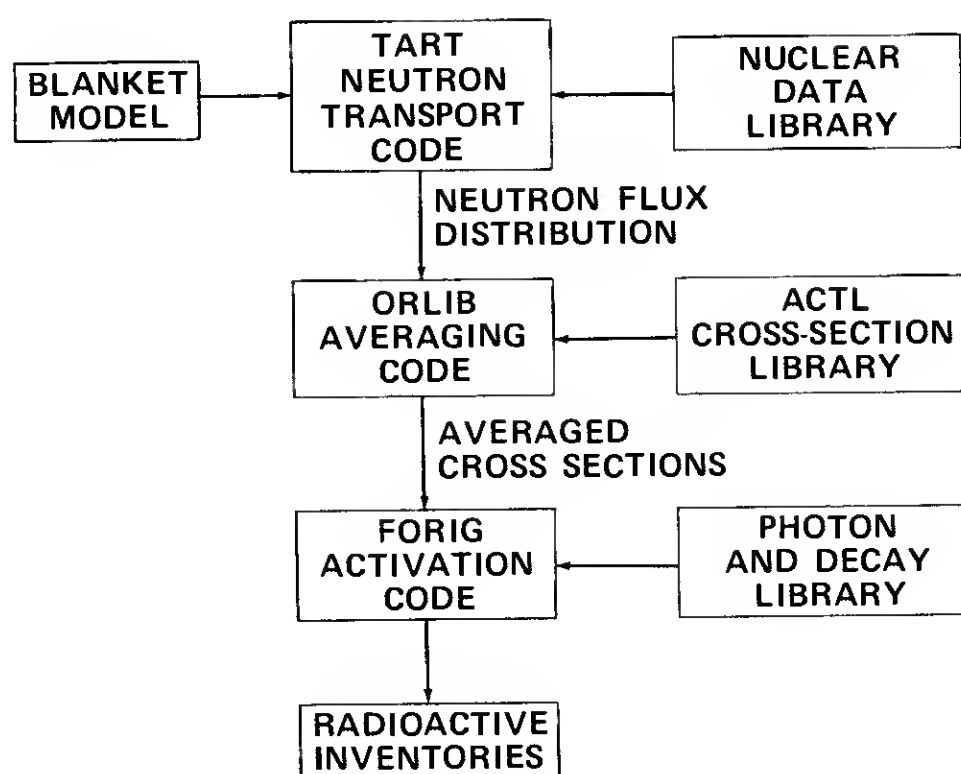


Fig. 1. Schematic diagram of LLNL neutronics package.

TABLE 1
First-Wall Radioactivity Calculated with Different Neutronics Packages

UWMAK-I, SS 316 ^a				MARS, HT-9 ^b			
Nuclide	Activity (Ci) Calculated by		UW/LLNL	Nuclide	Activity (Ci) Calculated by		UW/LLNL
	UW (1974) ^c	LLNL (1982) ^d			UW (1982) ^e	LLNL (1982) ^d	
²⁸ Al	1.7×10^7	8.1×10^6	2.0	²⁸ Al	2.2×10^6	3.6×10^6	0.62
⁴⁹ V	3.4×10^6	3.2×10^6	1.1	⁵¹ Ti	7.2×10^4	3.4×10^5	0.21
⁵² V	5.5×10^7	4.2×10^7	1.3	⁴⁹ V	4.1×10^6	9.7×10^5	4.2
⁵³ V	2.2×10^6	2.5×10^6	0.92	⁵² V	1.9×10^7	1.6×10^7	1.2
⁵¹ Cr	1.3×10^8	1.1×10^8	1.2	⁵¹ Cr	7.1×10^7	4.8×10^7	1.5
⁵⁵ Cr	7.0×10^6	2.8×10^6	2.5	⁵⁵ Cr	4.8×10^5	7.6×10^5	0.63
⁵⁴ Mn	1.2×10^8	9.1×10^7	1.3	⁵⁴ Mn	3.5×10^7	3.3×10^7	1.1
⁵⁶ Mn	2.1×10^8	2.1×10^8	1.0	⁵⁶ Mn	1.7×10^8	1.5×10^8	1.1
⁵⁷ Mn	2.0×10^6	2.5×10^6	0.81	⁵⁷ Mn	2.4×10^6	1.8×10^6	1.3
⁵³ Fe	1.0×10^6	3.5×10^5	2.9	⁵³ Fe	1.2×10^6	2.7×10^5	4.6
⁵⁵ Fe	7.0×10^8	6.2×10^8	1.1	⁵⁵ Fe	2.7×10^8	1.9×10^8	1.4
⁵⁷ Co	5.2×10^7	(Omitted)	---	⁵⁷ Co	8.4×10^5	(Omitted)	---
⁵⁸ Co	1.4×10^8	1.3×10^8	1.1	⁵⁸ Co	2.6×10^6	1.2×10^6	2.3
^{58m} Co	6.1×10^7	6.7×10^7	0.91	^{60m} Co	5.5×10^5	1.4×10^5	3.9
⁶⁰ Co	2.4×10^7	5.3×10^6	4.5	⁹² Nb	6.2×10^{-5}	1.1×10^6	6×10^{-11}
^{60m} Co	2.2×10^7	7.3×10^6	3.03	⁹¹ Mo	6.2×10^5	1.5×10^5	4.1
⁶¹ Co	4.0×10^5	5.1×10^5	0.78	⁹⁹ Mo	2.6×10^6	3.8×10^6	0.69
⁶² Co	3.1×10^5	2.5×10^5	1.2	¹⁰¹ Mo	3.1×10^5	9.9×10^5	0.32
⁵⁷ Ni	5.5×10^6	5.6×10^6	0.98	^{99m} Tc	2.6×10^6	3.3×10^6	0.79
⁹⁹ Mo	(Omitted)	9.4×10^6	---	¹⁰¹ Tc	3.1×10^5	9.9×10^5	0.32
¹⁰¹ Mo	(Omitted)	1.6×10^6	---	¹⁸¹ W	1.4×10^6	1.0×10^6	1.4
^{99m} Tc	(Omitted)	8.3×10^6	---	¹⁸⁵ W	2.2×10^6	2.8×10^6	0.77
¹⁰¹ Tc	(Omitted)	1.6×10^6	---	¹⁸⁷ W	4.3×10^5	1.7×10^6	0.25
Total	1.5×10^9	1.3×10^9	1.11	Total	5.9×10^8	4.7×10^8	1.26

^aAfter 10 full-power years at 1.25 MW/m².

^bAfter 2 full-power years at 5.0 MW/m².

^cUniversity of Wisconsin data from the UWMAK-1 report (Ref. 5), based on the ANISN-ENDF/B-III neutronics package. The reason for the long irradiation time (10 yr) is that Ref. 5 does not disaggregate the inventories by isotope for a 2-yr irradiation time.

^dData calculated at LLNL for this study, using the TART/ACTL neutronics package.

^eUniversity of Wisconsin (UW) data (Ref. 29), based on the ANISN-ENDF/B-IV neutronics package.

of the unlisted MPCs were estimated by analogy to listed isotopes. Table II indicates (a) that use of the "default" values of MPCs for isotopes not listed in the regulations produces contributions that misleadingly dominate the BHPs for all the materials shown except stainless steel, and (b) that the use of estimated MPCs in the past was able to reduce this problem but did not eliminate it.

IV.A.3. Dispersion Models

All dispersion and dose calculations are based on a release of 1-h duration, commencing 30 min after the postulated accident terminates fusion power generation. (The results are relatively insensitive to changes in these assumptions.)

The model for atmospheric dispersion of released radionuclides is a simple Gaussian plume similar to that employed in the Reactor Safety Study¹⁷ and in previous studies of doses from postulated fusion-reactor accidents.^{2,3,19,20} The model accounts for radioactive decay of the isotopes between reactor shutdown and the postulated release, as well as in transit. Production of radioactive daughters and their subsequent dispersion are accounted for. Ground deposition of radionuclides by the passing plume is included (deposition velocity = 0.01 m/s). Building-wake effects are approximated very crudely by initializing the plume's characteristic dimensions to those of the reactor building. No detailed treatment of thermal plume rise or radioactive self-heating is

TABLE II
First-Wall BHPs Using Various MPCs: MARS*

Isotope	BHPs (m ³) Using		
	Calculated ^a MPCs	Default ^b MPCs	Estimated ^c MPCs
Al-2024 wall	3 × 10 ¹⁶ (total)		
²⁷ Mg	2 × 10 ¹⁴	9 × 10 ¹⁷	3 × 10 ¹⁵
²⁸ Al	4 × 10 ¹³	4 × 10 ¹⁷	1 × 10 ¹⁵
SS 316 wall	9 × 10 ¹⁶ (total)		
⁴⁹ V	4 × 10 ¹³	1 × 10 ¹⁶	6 × 10 ¹²
⁵⁷ Mn	7 × 10 ¹¹	2 × 10 ¹⁶	6 × 10 ¹³
^{60m} Co	8 × 10 ¹¹	1 × 10 ¹⁵	5 × 10 ¹²
⁵⁷ Ni	4 × 10 ¹⁴	1 × 10 ¹⁵	1 × 10 ¹⁴
Nb—1 Zr wall	7 × 10 ¹⁶ (total)		
^{92m} Nb	5 × 10 ¹⁶	5 × 10 ¹⁸	1 × 10 ¹⁵
TZM wall	7 × 10 ¹⁶ (total)		
⁸⁹ Zr	6 × 10 ¹⁴	6 × 10 ¹⁶	6 × 10 ¹⁶
^{92m} Nb	2 × 10 ¹⁵	1 × 10 ¹⁷	4 × 10 ¹³
⁹⁶ Nb	2 × 10 ¹⁵	1 × 10 ¹⁷	1 × 10 ¹⁷
¹⁰¹ Mo	4 × 10 ¹⁴	8 × 10 ¹⁷	3 × 10 ¹⁵
¹⁰¹ Tc	2 × 10 ¹⁴	8 × 10 ¹⁷	3 × 10 ¹⁵

*Two full-power years at 5.0 MW/m².

^aCalculated using the ICRP models (see Ref. 23).

^b"Default" MPC = 10⁻¹⁰ Ci/m³ (see text).

^cSee Ref. 14.

included; the plume simply is assumed to originate at a height of 40 m above the ground.

IV.A.4. Dosimetry

Three routes of radiation exposure to individuals in the path of the plume of radionuclides are considered: inhalation from the passing plume and subsequent internal irradiation by the inhaled radionuclides, external irradiation by nuclides in the plume, and external irradiation by nuclides deposited on the ground by the plume. The internal-dose calculation accounts for all decay modes. The external-dose calculation includes only gamma radiation. (Most previous work in reactor-hazard assessment, including the Reactor Safety Study, also has ignored the external beta dose. Including it would increase the critical doses given below by 0 to 55% for all cases except silicon carbide (SiC), for which the increase would be ~400%.)

The amount of material inhaled is based on a breathing rate of 3.5 × 10⁻⁴ m³/s. This is the figure given by the ICRP for the standard man under conditions of light activity. The calculation of the resulting doses employs the physiological models and data of the ICRP (Ref. 31) and nuclear data from the *Table of Isotopes*,³² taking into account biological excretion, radioactive decay, and the effects of all

members of decay chains. The models and data for these dose calculations are described in detail in Ref. 33.

Although the models and data used are capable of determining the dose to any organ, only whole-body doses were calculated for the present purpose of generating a relative-hazard index. The whole-body dose was calculated, however, in two forms: the "critical dose" (identified in the Reactor Safety Study as the best predictor of early fatalities and acute radiation illnesses), amounting to all the dose delivered in the first week after exposure commences, plus half the dose delivered in the 8th through 30th days; and the 30-yr dose commitment resulting from the exposure.

The model for calculating the doses from external irradiation draws on the DOSFACTER (Ref. 34) and EXREM-III (Ref. 35) codes from the Oak Ridge National Laboratory, with modifications described in Ref. 33. The external dose calculation is based on the assumption that the individual remains in the contaminated area only during the passage of the plume; a longer stay would increase the dose received from ground-deposited radionuclides. Both cloud- and ground-dose calculations are integrated over the assumed distribution of radionuclides in the air or on the ground. This is a more rigorous procedure than multiplying the infinite-cloud or infinite-plane

doses by some overall correction factor, as was done in the Reactor Safety Study. The ground-dose calculation includes a shielding factor of 0.7.

We define our indices of relative hazard in terms of the critical dose and dose commitment to the whole body of an individual standing in the open, on the plume centerline, at a distance of 1 km from the postulated release. Because we are not concerned, under this definition, with the dose to a whole population, we are not obliged to deal with the complexities and ambiguities of evacuation assumptions and average shielding factors for a populace dispersed over various types of buildings and vehicles. Similarly, for purposes of determining an index of *relative* hazard (as opposed to trying to calculate an absolute figure for expected or maximum dose), it is unnecessary to struggle with averaging weather conditions properly or with picking the very worst case. We have simply chosen a somewhat typical weather condition—Pasquill class D atmospheric stability, no inversion, 5 m/s wind speed—for which the Gaussian plume model and finite-cloud dosimetry corrections are particularly straightforward and give reasonably accurate results.

IV.B. Results of Comparisons

We present here the results of applying the models just described to calculate radioactive inventories, BHPs, and off-site dose potentials for several combinations of fusion-reactor design and structural material.

IV.B.1. Cases Analyzed

The primary reactor design examined here is one developed in the MARS, a 1550-MW(electric) tandem mirror using lithium-lead ($\text{Li}_{17}\text{Pb}_{83}$) as the coolant/breeder.³⁶ Calculations based on this design were performed for the baseline first-wall material, HT-9 ferritic steel, and for six alternative materials: 316 SS; alloys of molybdenum, niobium, vanadium, and aluminum; and SiC ceramic.

These materials choices, intended only to explore to first order the hazard potentials associated with conceivable materials alternatives, are not necessarily all realistic in terms of material/coolant compatibility and in terms of the assumption of one-to-one substitutability, on a volume basis, of the alternative materials in the reference first wall. Indeed, some analysts consider it doubtful that aluminum alloys will be able to meet the requirements of service in a fusion-reactor first wall at all, and SiC is even more uncertain in this respect. On the other hand, both materials have their proponents,^{37,38} and both have been included here to help illustrate the full range of options that might exist.

To display the effects of large differences in the

reactor design itself, calculations were also performed for the UWMAK-I tokamak with two of the same first-wall-material alternatives—HT-9 and SS 316. The UWMAK-I was chosen both for comparability with the numerous previous studies that have calculated hazard indices for it and to provide the maximum contrast between an early and a contemporary design.

In addition to calculating hazard indices associated with the activated materials in fusion-reactor first walls, indices also were calculated for the "active" tritium inventories (tritium other than that held in cold storage as an operating reserve) and for the activated corrosion products circulating in the liquid-lithium coolant/breeder of a tokamak reactor. (Neither corrosion-product inventories nor lead activation have yet been calculated for the MARS design.) Finally, as another yardstick for comparison, the same BHP and dose models (with the same weather) were applied to the worst-case release postulated in the Reactor Safety Study for a pressurized water fission power reactor.

The relevant characteristics of the two fusion-reactor designs and the detailed compositions of the seven first-wall materials are given in Table III. The only elaboration needed on the information in the table has to do with the nature and extent of impurities in the SiC. Impurities tend to dominate the activation hazards of this material, and whether one must expect such contaminants at the level of 1 atomic part per million (appm) or at 10 or 100 appm is a matter of some controversy. In the end, however, the difference in these figures may be overwhelmed by the fraction of the first wall and associated structure for which ceramics turn out not to be usable at all. We have carried out our calculations for pure SiC in the first wall, permitting the reader to account for any postulated level of contamination or substitution by adding the appropriate fractions of the hazard indices of (say) steel.

IV.B.2. Comparisons of Indices and Materials

Table IV displays four different indices of potential hazard for the seven first-wall materials examined for the MARS design: the first-wall activity in curies; the associated BHP based on calculated (as opposed to default or estimated) MPCs; and the two "dose potentials" defined as the critical dose and 30-yr dose commitment to an individual 1 km from the postulated release of 100% of the first-wall material. (The weather conditions and other details of the calculation of these dose potentials were provided above.)

Our use of full first-wall dose potentials in Table IV and elsewhere in this paper is not intended to imply that a 100% release of the first-wall inventory is *likely* for any candidate wall material or

TABLE III
Material Compositions* and Reactor Configurations

HT-9 ^a :	Density = 7.78 g/cm ³ 0.20% C, 0.40% Si, 0.02% P, 0.02% S, 0.30% V, 11.5% Cr, 0.55% Mn, 85.0% Fe, 0.50% Ni, 1.0% Mo, 0.50% W
SS 316 ^a :	Density = 7.97 g/cm ³ 0.06% C, 0.007% N, 0.46% Si, 0.03% P, 0.01% S, 0.04% Ti, 16.7% Cr, 1.43% Mn, 64.44% Fe, 0.03% Co, 13.9% Ni, 0.06% Cu, 2.84% Mo
TZM ^a :	Density = 10.2 g/cm ³ 0.01% C, 0.50% Ti, 0.02% Fe, 0.01% Ni, 0.08% Zr, 99.4% Mo
Nb—1 Zr ^b :	Density = 8.55 g/cm ³ 0.75% Zr, 99.25% Nb
V—20 Ti ^c :	Density = 5.8 g/cm ³ 0.02% C, 0.05% N, 0.05% O, 0.004% Al, 0.03% Si, 0.01% P, 20.0% Ti, 79.79% V, 0.01% Fe, 0.008% Mo, 0.003% Ta, 0.01% W
Al-2024 ^d :	Density = 2.8 g/cm ³ 2.6% Mg, 90.3% Al, 0.48% Si, 0.086% Cr, 0.88% Mn, 0.48% Fe, 4.89% Cu, 0.23% Zn
SiC ^e :	Density = 3.24 g/cm ³ 30% C, 70% Si (see text about impurities)
UWMAK-I:	Tokamak reactor, major radius = 13 m, plasma radius = 5 m (calculations use a cylindrical approximation); thermal power = 5000 MW; electrical power = 1500 MW; neutron wall loading: 1.25 MW/m ² × 2830-m ² wall area; first-wall thickness = 0.25 cm, equivalent thickness = 0.4 cm; first-wall volume = 11.3 m ³ ; coolant/breeder: lithium
MARS:	Mirror reactor, length = 150 m, plasma radius = 0.4 m; thermal power = 4400 MW; electrical power = 1550 MW; neutron wall loading: 5 MW/m ² × 566-m ² wall area; first-wall thickness = 0.2 cm, equivalent thickness = 0.3 cm; first-wall volume = 1.70 m ³ ; coolant/breeder: Li ₁₇ Pb ₈₃

*Elemental compositions given in percent by weight.

^aData from Ref. 20.

^bDensity estimated from that of niobium. Composition data from Ref. 14.

^cComposition data from Ref. 20.

^dDensity estimated from that of aluminum. Composition data from Ref. 14.

^eDensity estimated by assuming face-centered cubic lattice structure with lattice parameter of 4.35 Å. Composition assumes 1:1 atomic ratio of carbon and silicon.

reactor design. For some design-material combinations, such a release may be inconceivable; making it so was a primary motivation of the choice of lithium-lead rather than liquid lithium as the coolant/breeder in the MARS design.^d For other design-material combinations, it may happen that larger releases (involving material from the rest of the blanket as well as from the first wall) cannot be completely ruled out. The intractable and vexatious topic of plausible release fractions is taken up in detail below.

Suffice it to say for now that we have based our dose potentials on first-wall inventories because this material represents the most concentrated structural

radioactivity in the reactor and, except for structural corrosion products in the coolant, is also the least well protected from release. Its vulnerability derives both from its high surface-to-volume ratio (promoting chemical attack and volatilization) and from its being subjected to more severe conditions (compared to what the rest of the structure suffers) both in normal operation and in accidents.

The dose potentials based on the full first-wall inventories are of course less illuminating about relative hazard than information about expected doses from realistic accident sequences would be—if only such information were available—but they are more illuminating by far than either numbers of curies or BHPs. They have the particularly attractive feature of telling one at a glance what a 10, 1, 0.1, or 0.01% release of first-wall material would amount to, enabling analysts with different views about the likelihood of releases of different sizes from various

^dThe adequacy of the lithium-lead choice for this purpose remains to be verified in detail; no doubt the degree of protection varies somewhat with the associated choice of first-wall material. See the discussion in Sec. V.

TABLE IV
Comparison of Indices of Potential Hazard for MARS

First-Wall ^a Material	Activity (10 ⁶ Ci)	BHP (10 ⁶ km ³)	Dose Potential (rem)	
			Critical	30 yr
HT-9	460	50	470	1100
SS 316	560	90	890	1600
TZM	940	70	1200	1600
Nb—1 Zr	540	70	2300	3800
V—20 Ti	280	30	410	700
Al-2024	290	30	1100	1100
SiC	280	0.4	0.34	0.35

^aFirst wall, after 2 full-power years at 5 MW/m². All wall materials occupy equal volume (see text).

first-wall/coolant combinations to draw their own conclusions about the relative and absolute hazards.

The comparisons of indices in Table IV suggest some interesting conclusions. Activity correlates at least roughly with critical-dose potential for the steels and the molybdenum and vanadium alloys, but not for niobium alloy, aluminum, and SiC. The BHPs are in line with critical-dose potentials for the two steels but for little else. The 30-yr-dose potentials, indicative of latent-cancer hazards, show less overall variation among materials than do the critical-dose potentials, which relate to early deaths and illnesses. (For example, TZM:SS 316:HT-9 = 1:0.7:0.4 for the critical dose but 1:1:0.7 for the 30-yr dose.) The ferritic steel, HT-9, is a remarkably good performer with respect to critical-dose potential, approaching vanadium alloy in this respect. Aluminum, on the other hand, is surprisingly bad on both dose potentials; its reputation as a low activation material does not seem justified in terms of potential hazard from reactor accidents. Silicon carbide, the only ceramic examined, is indeed in a class by itself, and would remain so even if contaminated to as much as a few percent by any of the alloys listed.

Table V shows the individual isotopes that account for the largest contributions to the dose potentials associated with the seven MARS first walls. In four of the seven cases, a single isotope accounts for 71% or more of the critical-dose potential. Table VI indicates the precursors and reactions that account for the major contributors to the dose potentials. The percentage abundances of the precursors, also shown in Table VI, offer little cause for optimism about reducing reactor-accident hazard potential by elemental or isotopic tailoring.^e

^eHazard potentials associated with the long-lived activation products that dominate waste-management considerations are a different matter. See, e.g., Ref. 39.

IV.B.3. Comparisons of Reactors and Release Types

Table VII compares some hazard indices from Table IV, based on the full activation-product inventories of two MARS first walls (HT-9 ferritic steel and 316 SS), with indices we have calculated analogously for several other circumstances: the full inventories of UWMAK-I first walls of the same materials; the full inventory of activated stainless steel corrosion products in a liquid-lithium-cooled tokamak; the active tritium inventories in the MARS and UWMAK-I designs; and the worst-case accidental release postulated for a pressurized water fission reactor in the NRC's 1975 study.¹⁷

In the case of the MARS-UWMAK first-wall comparison, the various hazard indices give a consistent impression, which holds for both materials: For the same rated output, the MARS design reduces the activation-hazard indices by nearly a factor of 2 compared to the early tokamak design.

Isotopic compositions of corrosion products were available for neither the MARS nor the UWMAK-I design, so an indicative worst-case figure for this type of inventory was taken from Piet's study²⁰ of the Starfire tokamak design. (The liquid-lithium/stainless steel combination produced by far the highest corrosion-product inventory of any of the several coolant/metal combinations Piet analyzed.) The associated hazard indices suggest that the release of coolant-borne corrosion products could produce nonnegligible consequences.

The active tritium inventories shown in Table VII range over two and one-half orders of magnitude, and the associated hazard indices clearly reveal the benefit of designing to the lower figures. Table VII also indicates that release of ~1% of the first-wall activity would produce hazards comparable to those from releasing all of the active tritium inventory in the base-case MARS design.

TABLE V
Important Contributors to Dose Potential for MARS*

Isotope	Half-Life	Inventory (Ci)	Dose Potential (rem)	
			Critical	30 yr
HT-9 ^a		4.6×10^8	470	1100
⁵⁵ Fe	2.6 yr	41%	3%	43%
⁵⁶ Mn	2.6 h	32%	49%	20%
⁵⁴ Mn	303 days	7%	37%	28%
SS 316		5.6×10^8	890	1600
⁵⁵ Fe	2.6 yr	28%	1%	24%
⁵⁶ Mn	2.6 h	24%	23%	13%
⁵⁸ Co	71.3 days	12%	45%	33%
⁵⁴ Mn	303 days	6%	21%	20%
TZM		9.4×10^8	1200	1600
⁹⁹ Mo	66.7 h	40%	71%	56%
^{99m} Tc	6.9 h	35%	6%	4%
⁹⁵ Nb	35 days	2%	8%	17%
^{92m} Nb	10.2 days	2%	5%	6%
⁹⁶ Nb	23.4 h	1%	4%	4%
Nb—1 Zr		5.4×10^8	2300	3800
^{92m} Nb	10.2 days	86%	87%	76%
⁹⁵ Nb	35 days	5%	11%	19%
V—20 Ti		2.8×10^8	410	700
⁴⁸ Sc	1.8 days	17%	76%	46%
⁴⁵ Ca	165 days	3%	4%	28%
⁴⁶ Sc	83.9 days	2%	18%	24%
Al-2024		2.9×10^8	1100	1100
²⁴ Na	15.0 h	46%	97%	96%
⁵⁴ Mn	303 days	1%	1%	2%
SiC		2.8×10^8	0.34	0.35
²⁹ Al	6.5 min	5%	32%	31%
²⁷ Mg	9.5 min	1%	17%	17%
³¹ Si	2.6 h	0.3%	27%	26%
²⁴ Na	15.0 h	0.003%	21%	20%

*First wall, 2 full-power years at 5 MW/m².

^aInventory for HT-9 differs slightly from Table I, reflecting small design changes in the blanket and shield structures.

In general agreement with earlier work,^{2,3} the comparison of hazard indices for the fusion first-wall activation products and the worst-case fission-reactor release shows only a modest advantage for fusion—less than half an order of magnitude—if the worst fusion case is chosen. The MARS design with HT-9 first wall, on the other hand, does better than fission by a factor of 7 or 8 based on these dose potentials.

Of course, the indices as presented in Table VII take no account of the relative probabilities for release of the inventories indicated. The PWR-1A fission release was assigned a probability of 4×10^{-7} /reactor year in the NRC study,¹⁷ although some analysts

would give a much higher figure and others a much lower one. The present state of fusion technology is not conducive to estimating such probabilities even roughly, but the nature of the materials is enough to suggest that the tritium is more readily released than the corrosion products and the corrosion products more readily released than (initially) solid structural materials. If one supposes that fusion-reactor accidents severe enough to release most of the tritium and much of the corrosion-product inventory are conceivable, it remains clear from Table VII that the fraction of first-wall material that can be released makes an enormous difference in the total

TABLE VI
Reactions Leading to Important Contributors to Dose Potential*

Isotope	Major Transmutations	Natural Abundance of Precursor (%)
HT-9 and SS 316		
^{54}Mn	$^{54}\text{Fe}(n,p); ^{55}\text{Mn}(n,2n)$	5.8; 100
^{56}Mn	$^{56}\text{Fe}(n,p); ^{55}\text{Mn}(n,\gamma)$	91.8; 100
^{55}Fe	$^{54}\text{Fe}(n,\gamma); ^{56}\text{Fe}(n,2n)$	5.8; 91.8
^{58}Co	$^{58}\text{Ni}(n,p)$	68.3
TZM		
^{92m}Nb	$^{92}\text{Mo}(n,p)$	15.9
^{95}Nb	$^{95}\text{Mo}(n,p)$	15.7
^{96}Nb	$^{95}\text{Mo}(n,p)^{95}\text{Nb}(n,\gamma)$	15.7
^{99}Mo	$^{98}\text{Mo}(n,\gamma); ^{100}\text{Mo}(n,2n)$	23.8; 9.6
^{99m}Tc	$^{98}\text{Mo}(n,\gamma)^{99}\text{Mo} \xrightarrow{\beta^-} 66 \text{ h}$	23.8
Nb-1 Zr		
^{92m}Nb	$^{93}\text{Nb}(n,2n)$	100
^{95}Nb	$^{94}\text{Zr}(n,\gamma)^{95}\text{Zr} \rightarrow$	17.4
	$^{96}\text{Zr}(n,2n)^{95}\text{Zr} \xrightarrow{\beta^-} 65 \text{ days}$	2.8
V-20 Ti		
^{48}Sc	$^{51}\text{V}(n,\alpha)$	99.7
^{45}Ca	$^{48}\text{Ti}(n,\alpha)$	73.7
^{46}Sc	$^{46}\text{Ti}(n,p)$	8.2
Al-2024		
^{24}Na	$^{27}\text{Al}(n,\alpha)$	100
Sic		
^{29}Al	$^{29}\text{Si}(n,p)$	4.7
^{27}Mg	$^{30}\text{Si}(n,\alpha)$	3.1
^{31}Si	$^{30}\text{Si}(n,\gamma)$	3.1

*MARS first wall, 2 full-power years at 5 MW/m².

hazard. It is to this crucial question of the release fractions from solid structural material that we now turn.

V. THE PROBLEM OF RELEASE FRACTIONS

The foregoing comparison of reactor designs and materials, emphasizing dose potentials based on the total inventories of the first walls considered, has the merit that these hazard indices have been self-consistently computed. The units in which these indices are expressed, moreover—rems of whole-body radiation dose 1 km from the reactor—facilitate an intuitive grasp of the meaning of the results. On the other hand, there is a danger that findings expressed in this way will be interpreted by some as measures of outcomes that are equally plausible for the various combinations of designs and materials examined, rather than as hazard-potential *inventories*, the plausibly releasable fractions of which may differ drastically from case to case. Clearly, then, it would

be preferable by far to weight these indices of hazard potential by multiplying them by the fractions of this potential that plausibly could be released in conceivable accidents. Alas, this prescription is much more easily described than executed.

In this section, we review briefly (a) the mechanisms germane to the release of activated structural material, (b) the attempts that have been made in previous fusion-reactor safety studies to establish plausible release fractions, and (c) the inadequacies of the applicable bodies of theory and experimental data, which, in our view, continue to make it impossible to establish such release fractions convincingly.

V.A. Release Mechanisms

Release of a significant fraction of the activated material in a fusion-reactor first wall requires, first of all, a mechanism for heating the material to temperatures above the normal operating temperature in the reactor. Such mechanisms include: (a) loss of

TABLE VII
Hazard Indices for Different Reactors and Release Types

Release Type and Reactor	Activity (10 ⁶ Ci)	BHP (10 ⁶ km ³)	Dose Potential (rem)	
			Critical	30 yr
100% of First Wall ^a				
SS 316				
UWMAK-I	970	200	1600	3000
MARS	560	90	890	1600
HT-9				
UWMAK-I	810	100	840	2000
MARS	460	50	470	1100
100% of Corrosion Products in Liquid-Lithium Coolant				
SS 316 Starfire ^b	13	3	13	48
100% of Active Tritium Inventory ^c				
MARS (base case)	20	0.1	5.4	9.2
MARS (low tritium)	6	0.03	1.6	2.8
UWMAK-1	240	1.2	65	110
Worst-Case Light Water Reactor Release				
PWR-1A ^d	1700	750	4000	7600

^aTwo full-power years at 5 MW/m².

^bSince isotopic compositions of corrosion products in liquid lithium were not available for UWMAK-I, values obtained by Piet (Ref. 20) for the Starfire tokamak with 316 SS first wall and liquid-lithium coolant were used here.

^cActive inventory includes all tritium except that in cold storage for operating reserve in case of breakdown of the tritium-extraction system. MARS inventory figures are from Ref. 36. UWMAK-I inventory figures are from Ref. 14. Doses assume release as HTO.

^dPressurized water reactor (PWR), assuming the release fractions given by Ref. 17 for a "PWR-1A" release. The results are scaled to 1.5 GW(electric), comparable to MARS and UWMAK-I.

heat-removal capability in the primary circuit; (b) loss of coolant flow; (c) loss of coolant; and (d) exothermic chemical reactions between coolant/breeder materials and air, water, or concrete. In cases (a), (b), and (c), the main energy source driving the heating would be afterheat in the activated structure itself; if the accident were accompanied by failure to shut down the fusion plasma, a much larger source term would come into play.

Given overheating, the main mechanisms for mobilizing activation products from structural materials would seem to be: (a) formation and vaporization of oxides or other relatively volatile compounds from overheated but still solid wall material; (b) the same from molten material; and (c) accelerated corrosion of wall material by overheated coolant, followed by volatilization of the corroded material from or with the coolant itself.

In all cases, release of the activated material to the environment requires, in addition to the foregoing, a breach in the reactor building and any containment envelopes internal to it. Such breaches could be caused by an external initiating event (aircraft impact, natural disaster, deliberate attack), by an internally generated projectile (massive magnet failure, failure of pressurized systems), by overpressure from fires, or by combinations of these.

V.B. Previous Studies

A number of authors have examined the loss-of-heat-removal, loss-of-coolant-flow, and loss-of-coolant possibilities for various designs and materials.^{20,40-42} Piet²⁰ has provided a particularly useful investigation of the margins, for various structural materials, between operating temperatures and the temperatures

that would be associated with different types of damages and release possibilities. The most troublesome structural material in this respect is molybdenum, which forms an oxide that volatilizes readily above ~ 900 K. Coupled with the high dose potential of activated molybdenum, this property seems to us to constitute a very significant liability mitigating against the use of high-molybdenum alloys.

For most other materials possibilities, achieving temperatures that could volatilize large quantities of activation products requires either failure to shut down the fusion plasma in an accident situation or a vigorous fire involving the coolant/breeder. Because it seems likely that highly reliable shutdown mechanisms for the fusion plasma can be provided, attention has focused on the second possibility, notably on lithium-air, lithium-water, and lithium-concrete fires.

V.B.1. Lithium Fires

Early theoretical work called attention to the very large stored energy in liquid lithium in the quantities envisioned for fusion reactors and to the high adiabatic flame temperature of this material's vigorous reactions with air, water, and concrete, in the range of 2100 to 2500 K. It was argued that the high atomic-oxygen concentrations occurring at these temperatures could eat through fusion-reactor first walls in matters of minutes by means of ablative oxidation.⁴

In the predecessor to the present paper,² Holdren assumed high mobilization rates of radioactivity from fusion-reactor first walls in lithium fires, based on evaporation of metallic elements from molten steel at 2300 K, catastrophic oxidation of niobium alloy brought on by melting of Nb_2O_5 above 1800 K, and rapid volatilization of MoO_3 above 1000 K. Kazimi⁴ criticized these estimates, arguing that his own work modeling lithium fires,^{3,19} plus the results of lithium-fire experiments at Hanford,⁴³ indicated that actual lithium-fire flame temperatures almost certainly will stay below 1500 K. (If that is so, lithium fires should neither melt steel nor catastrophically oxidize niobium. Kazimi did not dispute the possibility of high mobilization rates from molybdenum alloy.) More details of this work were published subsequently.⁴⁴

V.B.2. Oxidation and Volatilization

Piet's dissertation²⁰ contains a quite detailed review of the literature bearing on high-temperature oxidation and volatilization of alloys, in the context of the hazard indices for fusion reactors. He offers a "best estimate" for the volatilization rate of 316 SS in hot, dry air at 1270 K: 2×10^{-6} kg/m²·s, or $\sim 10^{-3}$ mm/h recession rate. Piet suggests a figure 100 times higher if catastrophic oxidation can occur

(as has been observed in this temperature range for Fe-Cr-Mo alloys of certain compositions).

V.C. Inadequacies in Theory and Data

Can one have confidence in any of the available estimates of the release fractions that are conceivable in lithium fires? We now think not. Our reasons are based on our own review—much extended since this paper's predecessor was written—of the literature of lithium fires and of the high-temperature mobilization of metals.

V.C.1. Theory and Experiments on Lithium Fires

The analytical modeling of lithium fires that has been performed at the Massachusetts Institute of Technology^{3,19,44} is impressive but as yet neither sufficiently comprehensive nor adequately verified experimentally to be completely persuasive. The assumptions used to derive the maximum flame temperature of 1500 K are by no means the most pessimistic possible about the quantity of lithium spilled, the area over which the pool spreads, and the available flow rate of air to sustain the combustion. The reports of this work acknowledge that larger spill areas increase the temperatures substantially and that the model does not predict very well the experimental results for "multi-species" lithium fires (which fusion-reactor lithium fires almost certainly would be).

The lithium-fire experiments for which we have seen documentation^{43,45} were rather modest in scope. The largest quantities of lithium involved were a few tens of kilograms, the largest pool areas on the order of a square metre. The test volumes were modest and the air-flow rates low. While it is certainly possible that the maximum temperatures observed in these tests will hold as well for larger fires, it is also possible that larger pool areas and higher air-flow rates will produce considerably more vigorous and hotter fires.

V.C.2. Theory of High-Temperature Volatilization

In most cases of conceivable interest, mobilization of structural metals will be dominated by the formation and volatilization of oxides rather than by the direct volatilization of pure metallic elements.⁴⁶ This being the case, the mobilization process can be characterized in terms of the following steps:

1. *Diffusion:* Oxygen molecules must diffuse through the carrier gas, oxide vapor layer, and any solid or liquid oxide layers on the surface of the metal; alternatively, the metal ions can diffuse through the oxide layers to the oxide-air interface.

2. *Adsorption:* Oxygen molecules must be physically adsorbed onto the surface of the metal or oxide. Then they may dissociate into atomic oxygen and be chemically adsorbed.

3. *Reaction*: The atomic oxygen and metal atoms must react to form the oxide.

4. *Vaporization*: The oxide molecule must vaporize and leave the surface.

5. *Convection*: The gaseous oxide must be convected through openings in the reactor structure without condensing.

These physical processes can be characterized by the rates at which they occur, and these rates are functions of pressure, temperature, and time. Since these processes occur in series, if one rate is much slower than the rest it will determine the overall reaction rate; this is called the "rate-limiting process." In mathematical terms, if $R_{i,max}$ is the theoretical maximum rate of the i 'th subprocess, then the theoretical overall reaction rate, R_{tot} , is given by

$$R_{tot} = [\sum_i (R_{i,max})^{-1}]^{-1}.$$

This equation indicates that, if one knows which subprocess is rate limiting, then the overall reaction rate can be approximated by calculating the rate of this subprocess.

The fundamental problem with this procedure is that the data needed to predict many of these rates are simply not available for the materials and conditions of interest here. Data for the processes that are most readily studied—gaseous diffusion, physical adsorption, and vaporization of individual compounds—often do not lend much insight about the overall problem, since the first two processes occur at very fast rates and, hence, are only of interest in cases of extreme volatility, and since vaporization is dependent on the combination of oxides that form. The formation of oxides, in turn, is dependent on solid-state diffusion and chemical reactions for which few data exist.

Theory can be used to predict the volatilization rates of pure metals for which basic thermodynamic data at high temperature are available. In the circumstances of present concern, however, neutron activation produces at least trace changes in chemical composition, which can influence volatilization behavior. Possibly these influences and the effects of alloying would be small in the cases of the TZM and Nb-1 Zr alloys, which are mostly molybdenum and niobium, respectively, and the main activation products of which are the same element as the parent material. All the other cases we are considering here involve complex alloys such as HT-9 or Al-2024, and the barriers to theoretical calculation become immense.

These barriers are of several kinds. First, since small changes in composition can yield large changes in material behavior, data should come from experiments on the alloys in question; but such data do not seem to be available except, in a very limited

sense, for SS-316. The results for simple systems cannot be applied to complex systems such as HT-9. Second, catastrophic oxidation must be considered in all systems containing molybdenum or vanadium, since small variations in these constituents are known to produce very large changes in volatility in simple systems. Third, it is extremely difficult to treat analytically the presence of lithium. Lithium is very corrosive, increasing the amount of metal released; on the other hand, it reduces the oxygen activity at the metal surface, making oxidation more difficult. Fourth, the preaccident reactor environment may change the behavior of materials in ways that are difficult to predict. For a period of 2 to 30 yr, the first wall will be exposed continuously to fluxes of high-energy neutrons up to 5 MW/m² and temperatures up to 800 K. Displacements caused by neutrons may alter the diffusion characteristics of the material significantly, and prolonged high temperatures may cause trace elements to migrate toward the surface at different rates.

Kofstad⁴⁶ states "It is clear that the oxidation behavior [of multicomponent alloys] is much too complex for a detailed analysis in terms of alloy and oxide phases, solid state reactions in the phases, etc. . ." We believe, moreover, that the further complications introduced by the preaccident reactor environment, the presence of lithium, and the generation and oxidation of trace elements created by neutron activation make it impossible to predict release fractions theoretically.

One complication poses special difficulties: There is good reason to believe that some elements may be released in quantities far out of proportion to their volume or weight fraction in the wall material, owing to differences in elemental diffusion rates, oxygen affinities, and so on. The problem this possibility poses for analysis and assessment is that some elements pose potential hazards hundreds of times greater than their fraction by weight would indicate. (This situation is illustrated in Table VIII, which is based on the calculations described in the foregoing sections.) If the elements that are preferentially released happen to be those that represent disproportionate shares of the hazard potential, then mobilizing a small fraction of the wall may release a much larger fraction of the potential hazard. The resulting necessity to predict differential mobilization of individual elements with great accuracy, if measures of the released hazard are to be accurate, far exceeds the present capabilities of theory.

V.C.3. Data on High-Temperature Volatilization

We have conducted an extensive literature search in the field of high-temperature oxidation of metals and have uncovered surprisingly few relevant results. One major difficulty is that investigations of the

TABLE VIII
Elemental Contribution to Weight and
Dose Potentials for MARS*

Element	Weight ^a (%)	Dose ^b (%)	Dose (%) Weight (%)
HT-9			
Phosphorus	0.020	0.45	22
Scandium	1.5×10^{-6}	0.30	2.0×10^5
Chromium	11.58	2.3	0.20
Manganese	0.90	87	97
Iron	84.5	3.6	0.043
Cobalt	0.0014	3.9	2800
Nickel	0.49	0.11	0.23
Niobium	0.0038	0.37	97
Molybdenum	0.98	1.9	1.9
Tantalum	0.0043	0.43	100
Tungsten	0.47	0.41	0.87
Rhenium	0.018	0.34	19
SS-316			
Phosphorus	0.030	0.15	5.0
Chromium	16.73	1.7	0.10
Manganese	1.67	44	26
Iron	64.28	1.7	0.027
Cobalt	0.056	47	840
Nickel	13.62	1.7	0.12
Niobium	0.011	0.59	54
Molybdenum	2.79	2.9	1.0
Technetium	0.028	0.24	8.4

*MARS first wall, 2 full-power years at 5 MW/m². All isotopes contributing more than 1 rem to critical dose potential are included.

^aPercent weight after irradiation (2 full-power years).

^bPercent of critical dose potential.

oxidation of alloys usually report only the weight loss of the sample over a period of time. Without reference to the oxide scale thickness and density, there is no way to tell how much of the weight loss is due to volatilization of the oxide. Furthermore, in many cases several different scales are formed, but the relative amounts and variation with time are not reported. Complex oxides called "spinels" often form, the composition of which is not accurately determined. Experimental conditions—usually very low oxygen pressures, moderately high temperatures, and low gas flow rates—tend to be far from those expected in a severe accident. The only alloy under consideration here that is treated specifically in the literature covered by our search is SS-316. Finally, no author we know of has directly measured the variable of interest: the composition of the volatilized material. Although this literature provides

some indication of the overall oxidation rate of metals, most of it is uninformative concerning the volatilization rates of individual constituents of alloys.

The two notable exceptions are experiments conducted at Hanford⁴⁵ and Idaho Falls.⁴⁷ In the lithium scoping studies done at Hanford, 45 kg of lithium was burned in a SS 316 pan in an unlimited supply of natural-humidity air. Liquid lithium at an initial temperature of 505 K ignited spontaneously with air, producing peak pool temperatures above 1310 K; the temperature of the pool remained above 1150 K for over 5 h. Lithium aerosols deposited on special disks on the floor were analyzed to determine the amount of each element volatilized.

Unfortunately, several aspects of the experiment cloud the results. First, although the supply of air was "unlimited," in practice this meant that the partial pressure of oxygen never dropped below 186 mbar. The flow rate of air through the room actually corresponded to that of a well-sealed building—hardly the case of a severe accident, where large inflows must be anticipated. Second, small quantities of ground metals were added to the lithium to simulate the presence of corrosion products. Rather than provide more information, the addition of these metals obscures the results, since it is impossible to tell which fraction of an element was volatilized from the wall and which originated from the ground metal. Although weight loss measurements might shed some light on this, the results reported are inconsistent; on one hand, it is reported that the pan weighed 4.3 kg less after the fire, but in the analysis of the results it is stated that 1 kg of metal was corroded. Finally, only oxide vapors scavenged by the lithium aerosols and deposited on the disks were analyzed, ignoring whatever material might have escaped the test chamber. Scavenging might be more effective for some elements than others.

The results of the experiment are stated in terms of concentration of elements in the aerosols in parts per million (ppm). By relating these back to the fraction of each element added or corroded, the authors conclude that manganese is the most releasable element, with vanadium a close second. If we assume that the metal corroded from the pan is released from the melt in the same proportion as the ground metal added to the lithium, then the corresponding release fraction of manganese from a SS 316 first wall would be ~0.005. It is difficult to put this number in perspective; to say, for example, whether it is a high or a low estimate. Actual lithium fires might generate higher or lower temperatures, and the air-flow rates could be hundreds of times greater (increasing oxygen activity and facilitating vaporization).

In experiments being conducted at Idaho Falls,⁴⁷ PCA (a stainless steel very similar in composition to

SS 316) is heated to temperatures of 1470 K in dry, flowing air. The vapors released during heating are condensed and sampled after 1, 21, and 74 h. Preliminary results indicate that molybdenum is released far out of proportion to its fraction by weight; the corresponding release fraction for a SS 316 first wall is ~ 0.03 . Manganese appears to be the second most releasable element, although far behind molybdenum. It is interesting that the Hanford and Idaho Falls experiments should give such different results; the differences may be accounted for by the presence of lithium, by the differences in temperatures, or by other factors.

V.D. What Can Be Concluded About Release Fractions?

Data that are truly applicable to the problem of high-temperature mobilization of metals from alloys of fusion-reactor interest are scanty. Theory is woefully inadequate. There is some experimental evidence for the proposition that releases of stainless-steel wall material in lithium fires producing temperatures below 1500 K would be below 1%, and that the same is true of overheated stainless steel in this temperature range but in the absence of lithium.

The same experiments indicate differential mobilization of elements that carry a disproportionate part of the potential hazard, however. Referring to Table VIII once again, if only 1% of the HT-9 first wall is released, but half of this release is manganese, then nearly 50% of the hazard is released. Similarly, if 1% of the SS 316 first wall is released, and only 6% of this is cobalt, then over 45% of the hazard is released.

Unfortunately, moreover, temperatures higher than 1500 K in large lithium fires cannot yet be ruled out convincingly. The experimental data base is too small and the range of parameters explored too narrow. A pessimist would point out also that the severe preaccident environment in fusion reactors could easily alter the properties of first-wall materials in ways that could enhance the mobilization rates of hazardous constituents. On the other side, the tendency of volatilized materials to plate out on cold surfaces before reaching the external environment might well reduce significantly the real possibilities for off-site doses.

VI. CONCLUSIONS AND RECOMMENDATIONS

The possibility of large releases of activated material from a molybdenum-alloy first wall under severe accident conditions is well established. Coupled with the high dose potential of activated molybdenum, this possibility constitutes an important liability of choosing molybdenum as a primary structural material. With respect to other candidate metals for first-wall applications, it cannot be shown

with confidence that large release fractions are possible—even in severe lithium fires—but neither can large release fractions be convincingly ruled out.

The dose potentials calculated here as an index of the hazard theoretically associated with the inventories of radioactivity in metal first walls, without reference to the probabilities of release, indicate that, *if large release fractions were possible*, off-site doses could fall in the range of 10 to 60% of those attributed by the same models to the worst-case fission-reactor release considered by the Reactor Safety Study. (The release fractions for the fission case have themselves been challenged as too high.⁴⁸) The differences in this respect among the alloys investigated amount to factors of 2 to 5, with the best performers being vanadium-titanium alloy and HT-9 ferritic steel. Silicon carbide, if feasible as a first-wall material, would offer an improvement of orders of magnitude in these indices, even if contaminated with metallic impurities to the extent of one percent or more.

Comparative studies of activation hazards, of the sort we have presented here, cannot be carried out simply by drawing on existing literature; a major reason is that the neutron-activation codes and data libraries used in different studies contain significant differences, preventing a literature review from distinguishing differences attributable to materials and designs from differences attributable to the calculational tools. It is hard to put great weight on the findings of any comparative-hazard studies—even those that consistently apply a single model and data set—unless and until the significant discrepancies in the major models and data libraries are all identified and explained. We believe there should be a coordinated effort to do this, beginning with the neutron-transport codes and cross-section libraries.

High priority should be given also to determining more convincingly the upper-limit temperatures producible in large-scale lithium fires at high air-flow rates. The needed experimental program should investigate at the same time the element-specific volatilization rates from candidate fusion-reactor structural materials exposed to these fires. Without this information, characterizing realistically the accident hazards of liquid-lithium-cooled fusion reactors will remain impossible.

Concern with the possibility of liquid-lithium fires has led to exploration of less reactive lithium-bearing compounds for coolant/breeder functions, notably lithium oxide, lithium aluminate, and the $\text{Li}_{17}\text{Pb}_{83}$ alloy in the MARS reactor design studied here. While there is no question that lithium oxide and lithium lead are less reactive than liquid lithium, more detailed studies are needed of the actual rates of energy release they can generate under conceivable accident conditions. Closer attention is also needed to the hazards associated with the activation of lead

in LiPb alloys, which we did not investigate in the present work.

Finally, the advantages of SiC in reducing the dose potentials associated with accidental release of first-wall materials are so striking that more extensive exploration of whether the use of this material is actually feasible seems well warranted.

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